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PATENT 1781-0221P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant:

BAOZONG, Zhao et al.

Conf.:

6648

Appl. No.:

09/828,924

Group:

1751

Filed:

April 10, 2001

Examiner: VIJAYAK

For:

ELECTRICALLY CONDUCTIVE POLYMERS

LETTER

Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450 November 18, 2003

Sir:

Under the provisions of 35 U.S.C. § 119 and 37 C.F.R. § 1.55(a), the applicant(s) hereby claim(s) the right of priority based on the following application(s):

Country

Application No.

Filed

SINGAPORE

200002026-3

April 11, 2000

A certified copy of the above-noted application(s) is(are) attached hereto.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fee required under 37 C.F.R. §§ 1.16 or 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

P.O. Box 747

DRN:trb

1781-0221P

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Attachment(s)

1781-221P 09/828,924 Baozong etal. BSKB (403)205-8000

REGISTRY OF PATENTS SINGAPORE

This is to certify that the annexed is a true copy of specification as filed for the following Singapore patent application.

Date of Filing

11 APRIL 2000

Application Number

200002026-3

Applicant(s) / Proprietor(s) of

Title of Invention

NATIONAL UNIVERSITY OF

SINGAPORE

Patent

ELECTRICALLY CONDUCTIVE

POLYMERS

SHARMAINE WU (Ms)
Assistant Registrar
for REGISTRAR OF PATENTS

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200002026-3

1 1 APR 2000

PATENTS FORM 1

Rule 19

SINGAPORE **PATENTS ACT** (CHAPTER 221) PATENTS RULÉS

The Registrar of Patents Registry of Patents

REQUEST FOR THE GRANT OF A PATENT
THE GRANT OF A PATENT IS REQUESTED BY THE UNDERSIGNED ON THE BASIS OF THE PRESENT APPLICATION

I. Title of Invention	ELECTRICALLY CONDUCTIVE POLYMERS						
II. Applicant(s) (See note 2)	(a) Name	National University of Singapore					
(See note 2)	Body Description/	Incorporated in Singapore					
	Residency Street Name & Number	10 Kent Ridge Crescent					
	City	Singapore					
	State	Singapore					
	Country	Singapore					
•	(b) Name						
	Body Description/ Residency						
	Street Name & Number						
	City						
	State						
	Country						
	(c) Name						
	Body Description/						
	Residency						
	Street Name & Number						
	City						
	State	·C					
-	Country						

200002026-3

III. Declaration of Priority (see note 3)	Country/Country Designated Filing Date Country/Country Designated				File no.				
	Filing Date Country/Country Designated	twi t			File no.	:			
	Filing Date								
IV. Inventors (See note 4) (a) The applicant(s) is sole/joint inventor (b) A statement on Portion 8 is/will be joint inventor.	(s). atents furnished.		х	Ye Ye			X	No No	
V. Name of Agent (if any) (S	See note 3)						. •		
VI. Address for Service (See note 6)		•	ck/Hse No it No/PO		ent Ridge O Box 10	;	Level No Postal Cod	ie	911101
		Str	eet Name		<u>.</u>	+			
			lding Name						
VII. Claiming an earlier filing date under section 20(3), 26(6) or 47(4). (See note 7)			olication No						
		Fili	ng Date		_,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,				
		[Please tick in the relevant space provided]: () Proceeding under rule 27(1)(a). Date on which the earlier application was amended =							
	_	or () Proceeding	und	er rule 27((1)(<i>b</i>).		_	· · · · · · · · · · · · · · · · · · ·

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VIII. Invention has been displayed at an International Exhibition (See note	Yes X No								
IX. Section 114 requirements (See note 9)	The invention relates to and/or used a micro-organism deposited for the purposes of disclosure in accordance with section 114 with a depository authority under the Budapest Treaty. Yes X No								
X. Check List	A. The application contains the following number of			of sheet(s):-		,			
(To be filled in by applicant or agent)	1. Req	uest		4		sheets			
en e	2. Des		9		sheets				
	3. Cla	im(s).		3	3				
,	4. Dra	wing(s).		2		sheets			
	5. Abs	tract.		1		sheets			
	B. The application as filed is accompanied by:-								
	1. Prio	ority docui	ment		L				
	2. Tra	nslation o	f priority document		L				
· ·	3. Stat	ement of l	Inventorship & right to grant		x				
	4. International Exhibition Certificate					製造			
X1. Signature(s)	Applicant	t (a)	Cylhoto	The same was harden to the					
(See note 10)	Date		10 April 2000	and the last					
	Applicant	t (b)	, .						
	Date				為這	法建立			
	Applicant	t (c)							
	Distr		1	5型公司 第	建築組				

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NOTES:

- 1. This form when completed, should be brought or sent to the Registry of Patents together with the prescribed fee and 3 copies of the description of the invention, and of any drawings.
- 2. Enter the <u>name and address of each applicant</u> in the spaces provided at paragraph II. <u>Names of individuals</u> should be indicated in full and the surname or family name should be underlined. <u>The names of all partners</u> in a firm must be given in full. The <u>place of residence of each individual</u> should also be furnished in the space provided. Bodies corporate should be designated by their <u>corporate name</u> and <u>country of incorporation</u> and, where appropriate, the <u>state of incorporation</u> within that country should be entered where provided. Where more than 3 applicants are to be named, the names and address of the fourth and any further applicants should be given on a <u>separate sheet</u> attached to this form together with the <u>signature of each of these further applicants</u>.
- 3. The declaration of priority at paragraph III should state the date of the previous filing, the country in which it was made, and indicate the file number, if available. Where the application relied upon in an International Application or a regional patent application e.g. European patent application, one of the countries designated in that application [being one falling under the Patents (Convention Countries) Order] should be identified and the name of that country should be entered in the space provided.
- 4. Where the applicant or applicants is/are the sole inventor or the joint inventors, paragraph IV should be completed by marking the 'YES' Box in the declaration (a) and the 'NO' Box in the alternative statement (b). Where this is not the case, the 'NO' Box in declaration (a) should be marked and a statement will be required to be filed on Patents Form 8.
- 5. If the applicant has appointed an agent to act on his behalf, the agent's name should be indicated in the spaces available at paragraph V.
- 6. An address for service in Singapore to which all documents may be sent must be stated at paragraph VI. It is recommended that a telephone number be provided if an agent is not appointed.
- 7. When an application is made by virtue of section 20(3), 26(6) or 47(4), the appropriate section should be identified at paragraph VII and the number of the earlier application or any patent granted thereon identified. Applicants proceeding under section 26(6) should identify which provision in rule 27 they are proceeding under. If the applicants are proceeding under rule 27(1)(a), they should also indicate the date on which the earlier application was amended.
- 8. Where the applicant wishes an earlier disclosure of the invention by him at an International Exhibition to be disregarded in accordance with section 14(4)(c), then the 'YES' Box at paragraph VIII should be marked. Otherwise the 'NO' Box should be marked.
- 9. Where in disclosing the invention the application refers to one or more micro-organisms deposited with a depository authority under the Budapest Treaty, then the 'YES' Box at paragraph IX should be marked. Otherwise the 'NO' Box should be marked.
- 10. Attention is drawn to rules 90 and 105 of the Patent Rules. Where there are more than 3 applicants, see also Note 2 above.
- 11. Applicants resident in Singapore are reminded that if the Registry of Patents considers that an application contains information the publication of which might be prejudicial to the defence of Singapore or the safety of the public, it may prohibit or restrict its publication or communication. Any person resident in Singapore and wishing to apply for patent protection in other countries must first obtain permission from the Singapore Registry of Patents unless they have already applied for a patent for the same invention in Singapore. In the latter case, no application should be made overseas until at least 2 months after the application has been filed in Singapore.

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Application Filing D	ate:	1	1		
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Fee received on	:	1	1		
Amount	:				

ELECTRICALLY CONDUCTIVE POLYMERS

Field of the invention

The present invention relates to a method of preparing electrically conductive polymeric material by treatment with a viologen salt and to electrically conductive polymeric articles formed thereby.

Description of related art

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In recent years electrically conductive polymers have been widely studied because of their potential important commercial applications. Such polymers include polypyrrole and its derivatives and polyaniline and its derivatives.

The aniline family of polymers is an example of one of the most widely studied electroactive polymers because of the ease of synthesis and the unusual nature of its electrical conductivity which can be varied over a wide range. Polyaniline (PANi) in its insulating base form is readily soluble in N-methylpyrrolidinone and can be cast into films or coated on different substrates. The insulating form can be converted to the conductive form by the following methods:

(1) Treatment with protonic acids

PANi in the emeraldine (50% oxidated) base state when treated with protonic acids shows an increase in electrical conductivity. The resulting conductivity is a strong function of the pH of the equilibrating solution. For example, when aqueous HCl is used at pH greater than 4, the PANi remains as the essentially insulating non-protonated form whereas at pH~0 (i.e. with 1 M HCl), the conductivity increases by 10 orders of magnitude (J. C. Chiang and A. G. MacDiarmid, *Synth. Met.* 13, 193 (1986)). In the latter case, the imine units of the PANi are protonated to give a N*/N ratio of about 0.5. In this form of doping, there is no change in the number of electrons in the PANi chains.

The electrical conductivity of the PANi treated with protonic acids depends not only on the pH of the equilibrating solution but also on the oxidation states of the PANi. For example, PANi in the leucoemeraldine (100% reduced) state shows only a small increase in conductivity when treated with protonic acids (J. C. Chiang and A. G. MacDiarmid, *Synth. Met.* 13, 193 (1986)).

(2) Charge transfer interaction with organic electron acceptors

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PANi in the emeraldine base state can undergo charge transfer interaction with organic electron acceptors such as tetrachloro-o-benzoquinone and 2,3-dichloro-5,6-dicyano-p-benzoquinone in acetonitrile (S. H. Khor, K. G. Neoh and E. T. Kang, *J. Appl. Polym. Sci.* 40, 2015 (1990)). An increase in electrical conductivity is achieved and the magnitude of the conductivity is dependent on the type of organic acceptors and acceptor concentration. The charge transfer interaction of emeraldine base with the organic electron acceptor is analogous to that of protonation by HCI, with the hydrogen atoms in the latter replaced by the partially dehalogenated halobenquinone rings. The maximum conductivity achieved is at least an order of magnitude lower than that achievable in protonic acid doping of emeraldine base.

PANi in the leucoemeraldine state undergoes oxidative doping by organic electron acceptors in acetonitrile solution which involves first the oxidation of the amine nitrogen and also the formation of the imine structure through hydrogen transfer from the amine nitrogen to the acceptor. The so-produced imine nitrogen is then in turn doped by the acceptor, as described above for PANi in the emeraldine state (E. T. Kang, K. G. Neoh, T. C. Tan, S. H. Khor, and K. L. Tan, *Macromolecules*. 23, 2918 (1990)). The maximum electrical conductivity obtained is also about an order of magnitude lower than that obtained from protonic acid doping of emeraldine.

It would be a significant advance in the art if a method of preparing an electrically conductive polyaniline material could be provided which would function irrespective of the oxidation state of the polyaniline. The polyaniline base material may be a substituted or unsubstituted polyaniline. It would be a further significant

advance in the art if a method could be provided which did not involve the use of acid or organic solvents such as acetonitrile.

It is accordingly an object of the present invention to overcome, or at least alleviate, one or more of the difficulties and deficiencies related to the prior art.

5 Summary of the invention

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Accordingly, in a first aspect of the present invention there is provided an electrically conductive polymeric article including a polymeric material capable of exhibiting electrical conductivity;

the polymeric material being rendered electrically conductive by treatment with a viologen salt.

It has surprisingly been found that the electrically conductive polymeric article according to this aspect of the present invention exhibits a high level of electrical conductivity. For example, electrically conductive polyaniline may be prepared with resistances (Rs) decreasing from approximately 10^{10} to approximately 10^{6} Ω /sq or below, irrespective of the oxidation state of the polyaniline material. Polyaniline material may be in the leucoemeraldine (0% oxidation state) or emeraldine (50% oxidation state) states or in any oxidation state between these two states.

Examples of polymeric materials capable of being rendered electrically conductive by treatment with a viologen include polymers derived from aromatic bases such as aniline and its derivatives and from heterocyclic bases such as pyrrole and its derivatives.

Prepared polymeric materials for inclusion in the polymeric articles of the invention include polyaniline and its derivatives and polypyrrole and its derivatives. Polyaniline and polypyrrole are particularly preferred polymeric materials.

The electrically conductive polymeric material may be formed in any suitable manner and may take any suitable shape. The polymeric may be in the

form of a film, e.g. a free standing film, a film coating, e.g. a thin film coating, or a powder.

In a preferred aspect the polymeric material may be deposited or supported on a suitable substrate. The substrate may be a fabric, polymeric matrix film or other construct. A polyethylene substrate, e.g. a low density polyethylene (LDPE) substrate may be used.

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The viologen salt utilised to dope the polymeric material may be of any suitable type. Viologen salts such as a dihalide salt of a viologen (1,1'-disubstituted 4,4'-bipyridinium dihalide) have been found to be particularly suitable.

The substituents on the bipyridinium molecule may be selected from alkyl or aryl groups. The alkyl groups may be substituted or unsubstituted C1 to C4 alkyl groups. The alkyl groups may be straight or branched chains. Substituents for the alkyl groups may be selected from a wide range of substituents including halogen, phenyl and substituted phenyl. Examples of preferred alkyl groups include methyl, ethyl, propyl, butyl and phenylmethyl (benzyl). The aryl groups may be substituted or unsubstituted aryl or heteroaryl.

Viologen salts may be provided as free compounds, shown below as I, or in the form of polymers, shown below as II, where:

R and R¹ are independently chosen from alkyl or aryl groups as indicated 20 above;

 R^2 is an alkylene group, preferably a C_2 to C_4 alkylene group;

X is an anion, preferably a halide ion; and

n is an integer greater than or equal to 2.

Particularly preferred viologens include ethyl viologen dichloride, benzyl viologen dichloride and poly(butyl viologen dibromide).

$$R-N$$
 $X N-R'$
 $X-$

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$$(II)$$

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In a preferred aspect of the present invention the polymeric material may be directly treated with a viologen to render it electrically conductive.

Accordingly, in a preferred aspect of the present invention there is provided a method of preparing an electrically conductive polymeric article, which method includes providing

> a polymeric material capable of exhibiting electrical conductivity; and a viologen;

contacting a surface of the polymeric material with the viologen for a time sufficient to permit the polymeric material to be rendered electrically conductive.

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Although not wishing to be bound by theory it is believed that when the polymeric material is treated with a viologen salt that the polymeric material undergoes oxidative doping and the halide anions are incorporated into the polymer. Surprisingly the method may be conducted at or above room temperature in the presence of air and preferably utilising an aqueous solution of the viologen salt. Thus the use of protonic acids or organic solvents may be avoided.

The rate of the reaction is dependent on the ease of the reduction of the viologen, e.g. a viologen dication, and the transfer of the anions to the polymeric material to form a doped and conductive form of the polymeric material.

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Pursuant to the present invention, the reactions may be carried out by contacting the surface of the polymeric material with an aqueous solution of viologen salt. The reactions may continue for a time sufficient to permit a substantial degree of oxidative doping of the polymeric material to be achieved.

The viologen salt may be selected from any suitable materials as discussed above. A viologen dihalide is preferred.

The method of contacting the surfaces of the polymeric material with the viologen may vary widely, depending on the state of the polymeric material and the method selected for the reactions. One simple and preferred method involves immersing the polymeric material in an aqueous solution of the viologen salt, preferably a viologen dihalide, more preferably in excess, with proper agitation. The preferred concentration of the solution is between 0.01 M and 0.5 M.

The method may be conducted at any suitable temperature, e.g. at a temperature between 0° and approximately 100°C. The method may be conducted in the presence of air and under ordinary room lighting.

The method may be carried out for a period of time which is dependent on the form of the polymeric material (coating, thick film or powder), type of viologen salt and concentration. For example, for a thin PANi coating and a monomeric viologen dichloride at a concentration of 0.1 M, the reaction is rapid and the film turns conductive within 5 mins. For thick PANi films, and using a similar viologen type and concentration, the period of the experiment is extended to 1 day to ensure completion.

After completion of the reaction, the polymeric material may, for example, be blotted dry between filter paper to remove the unreacted viologen salt. The change of the PANi from the insulating base state (brown and blue for emeraldine freestanding film and coating, respectively) to the conductive state (blue and green for freestanding film and coating, respectively) is visually apparent, and may be

confirmed by sheet resistance (Rs) measurement, X-ray photoelectron spectroscopy or UV-visible absorption spectroscopy. Figure 1 shows the sheet resistance (Rs in Ω /sq.) of emeraldine base free standing film treated in 0.24 M benzylviologen dichloride. Figure 2 shows the UV-visible absorption spectra of PANi coating on LDPE film treated in 0.024 M polybutylviologen dibromide.

EXAMPLES

The following specific examples are provided to illustrate this invention and the manner in which it may be carried out. It will be understood, however, that the specific details given in each example have been selected for the purpose of illustration, and are not to be construed as being limitations on the scope of the invention.

EXAMPLE 1

Polyaniline (in the emeraldine or 50% oxidation state) was synthesized via the oxidative polymerization of aniline using ammonium persulfate in 0.5 M H_2SO_4 (A.G. MacDiarmid et al. *Synth. Met.* 18, 285 (1987)). The polyaniline powder was undoped using excess 0.5 M NaOH and the polyaniline (emeraldine) base powder was dissolved in N-methyl pyrrolidinone (NMP). Freestanding film of 10-20 μm was cast from the polyaniline base (8% wt) in NMP solution. This film was immersed in a 0.24 M solution of benzyl viologen dichloride

in water. The pH of the solution was between 5 and 6 and the experiment was carried out at 25 °C in an open test tube. A sharp decrease in Rs was observed within the first hour. The change in Rs with time of treatment is shown in Figure 1.

The doping level as given by the N⁺/N ratio obtained is 0.4 after 24 h.

EXAMPLE 2

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A coating of polyaniline (in the emeraldine state) on low density polyethylene (LDPE) substrate was prepared by immersing a O_2 plasma pretreated LDPE film into a reaction mixture containing 0.10 M aniline and 0.025 M ammonium persulfate in 0.5 M H_2SO_4 for 2 h. The green polyaniline coated LDPE film was undoped in 0.5 M NaOH for 2 h and then washed with deionized water and pumped dry under reduced pressure. The base film was immersed in a 0.024 M poly(butylviologen dibromide)

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solution in water under ordinary room lighting. The change in the UV-visible absorption spectrum from that of emeraldine base to a conductive salt is illustrated in Figure 2.

EXAMPLE 3

Polyaniline freestanding (emeraldine) base film was synthesized as described in Example 1. Leucoemeraldine was prepared by treating the emeraldine base film with 10% hydrazine for 48 h. After treatment of the leucoemeraldine film (pale brown) with 0.12 M benzyl viologen dichloride at 25 °C and under ordinary room lighting for 24 h, the film has turned blue and Rs decreased from 10^{10} to 6×10^5 Ω/sg .

EXAMPLE 4

Polyaniline freestanding (emeraldine) base film was synthesized as described in Example 1. Treatment of the film with 0.12 M benzyl viologen dichloride at 5 °C was carried out for 24 h under ordinary room lighting. The Rs obtained was 3×10^5 Ω/sq .

EXAMPLE 5

Polyaniline freestanding (emeraldine) base film was synthesized as described in Example 1. Treatment of the film with 0.12 M benzyl viologen dichloride at 25 °C was carried out for 24 h in the absence of light. The Rs obtained was $4\times10^5~\Omega/\text{sq}$.

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EXAMPLE 6

Polypyrrole (PPY) freestanding film was electrochemically synthesized in a one-compartment cell with 0.1 M pyrrole, 0.1 M p-toluenesulfonic acid in acetonitrile containing 1% v/v water. The film was grown with a charge density of 10 Coulomb cm⁻² at 0~5° C (X. Zhang, E. T. Kang, K. G. Neoh, K. L. Tan, D. Y. Kim and C. Y. Kim, *J. Appl. Polym. Sci.* 60, 1996, 625). The PPY film was then undoped by treatment in 0.5M NaOH for 24h. This dried PPY film was immersed in 0.24 M aqueous solution of benzyl viologen dichloride for 24h at 25°C under ordinary room lighting. The film turned dark blue and Rs decreased from 10⁷ to 10⁴ o/sq.

Finally, it is to be understood that various alterations, modifications and/or additions may be made without departing from the spirit of the present invention as outlined herein.

CLAIMS

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1. An electrically conductive polymeric article including a polymeric material capable of exhibiting electrical conductivity;

the polymeric material being rendered electrically conductive by treatment with a viologen salt.

- 2 An electrically conductive polymeric article according to claim 1 wherein the polymeric material is chosen from polymers derived from aromatic bases and from polymers derived from heterocyclic bases.
- 3. An electrically conductive polymeric article according to claim 2wherein the polymeric material is chosen from polyaniline and its derivatives.
 - 4. An electrically conductive polymeric article according to claim 2 wherein the polymeric material is chosen from polypyrrole and its derivatives.
 - 5. An electrically conductive polymeric article according to claim 3 wherein the polymeric material is polyaniline.
- 15 6. An electrically conductive polymeric article according to claim 4 wherein the polymeric material is polypyrrole.
 - 7. An electrically conductive polymeric article according to Claim 5, wherein the polyaniline base material has an oxidation state between the leucoemeraldine (0% oxidation state) and the emeraldine (50% oxidation state).
- 20 8. An electrically conductive polymeric article according to Claim 1, wherein the polymeric material is in the form of a film, film coating, or powder.
 - 9. An electrically conductive polymeric article according to Claim 8, wherein the polymeric material is deposited on a suitable substrate.
- 10. An electrically conductive polymeric article according to Claim 1, wherein the viologen salt is a viologen dihalide.



- 11. An electrically conductive polymeric article according to claim 10 wherein in the viologen dihalide the substituents on the bipyridinium are chosen from substituted and unsubstituted alkyl and aryl groups.
- 12. An electrically conductive polymeric article according to claim 11 wherein the substituted and unsubstituted alkyl groups are chosen from C1 to C4 alkyl optionally substituted with halogen, aryl or substituted aryl.
 - 13. An electrically conductive polymeric article according to claim 12 wherein the substituted and unsubstituted alkyl groups are chosen from C1 to C4 alkyl and benzyl.
- 10 14. An electrically conductive polymeric article according to claim 10 wherein in the viologen dihalide the halide anions are chosen from chloride, bromide and iodide.
 - 15. An electrically conductive polymeric article according to Claim 1, wherein the viologen salt is present in a monomeric or polymeric form.
- 16. An electrically conductive polymeric article according to Claim 10, wherein the haloviologen material is selected from one or more of benzyl viologen dichloride and poly(butylviologen dibromide).
 - 17. An electrically conductive polymeric article according to Claim 5, wherein the resistance of the polyaniline base material, Rs, is reduced from approximately $10^{10} \Omega/\text{sq}$ to approximately $10^6 \Omega/\text{sq}$ or less.
 - 18. A method of preparing an electrically conductive polymeric article, which method includes

providing

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a polymeric material capable of exhibiting electrical conductivity; and a viologen salt;

contacting a surface of the polymeric material with the viologen salt for a time sufficient to permit the polymeric material to be rendered electrically



conductive.

- 19. A method according to Claim 18, wherein the polymeric material is a polyaniline which has an oxidation state between the leucoemeraldine (0% oxidation state) and the emeraldine (50% oxidation state).
- 5 20. A method according to claim 18 wherein the polymeric material is a polypyrrole.
 - 21. A method according to Claim 18, wherein the viologen salt is a viologen dihalide.
- 22. A method according to Claim 21, wherein the viologen salt is selected from one or more of viologen benzyl dichloride and poly(butylviologen dibromide).
 - 23. A method according to Claim 10, wherein the viologen salt is present in the form of an aqueous solution.
- 24. A method according to Claim 23, wherein the method is conducted at a temperature of 0° to approximately 100°C in the presence of air.
 - 25. A method according to Claim 10, wherein the rate of conversion of the polymeric material to a conducting state is varied by varying one or more of the type and concentration of the viologen salt, the oxygen concentration, temperature and exposure to light.
- 26. An electrically conductive polymeric article whenever prepared according to the method according to Claim 18.



ABSTRACT

An electrically conductive polymeric article including a polymeric material capable of exhibiting electrical conductivity;

the polymeric material being rendered electrically conductive by treatment with a viologen salt.

(Figure 1 is to be published)

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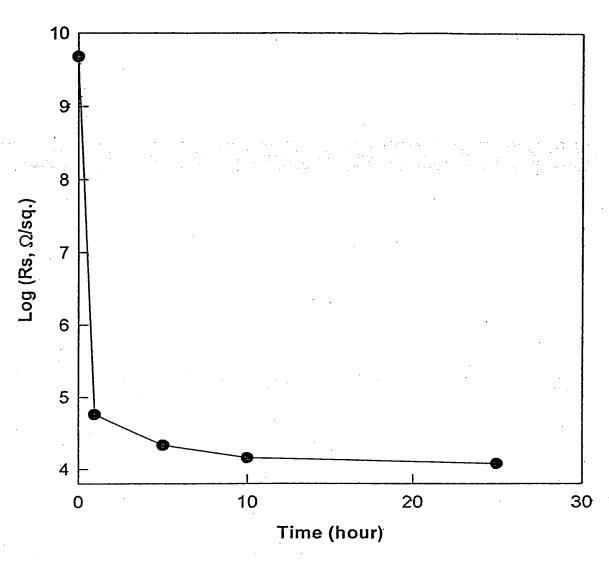


Figure 1

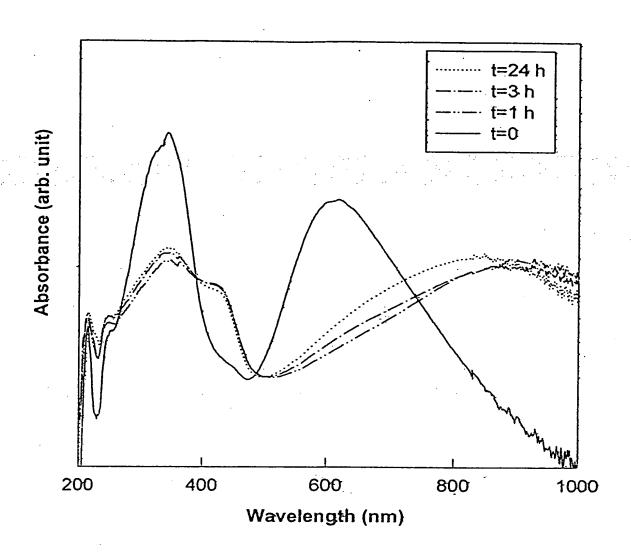


Figure 2